Doping Dependence of the Magnetic Resonance Peak in $YBa_2Cu_3O_{6+x}$

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We report inelastic neutron scattering experiments on the doping dependence of the energy and spectral weight of the sharp magnetic resonance peak in $YBa_2Cu_3O_{6+x}$. These measurements also shed light on the relationship between the magnetic excitations in the normal and superconducting states.

Magnetic excitations in high temperature superconductors have been intensively studied experimentally and theoretically for a number of years as they provide a direct and incisive probe of correlation effects in the cuprates. These efforts have been redoubled after the discovery of a sharp magnetic collective mode in $YBa_2Cu_3O_7$ by inelastic neutron scattering [1–3]. This mode is strongly coupled to superconductivity in this material; in fact, it is only present in the superconducting state and disappears in the normal state [1]. Two different mechanisms, with various modifications, have been proposed to explain this observation. First, it may be a consequence of the pileup of electronic states above the superconducting energy gap which compensates for the loss of states below the gap. Both a *d*-wave BCS gap function with strong Coulomb correlations [4–6] and the (non-BCS) gap function resulting from the interlayer pair tunneling model of superconductivity [7] can account for the sharpness of the mode in both wavevector $\mathbf{q}$ and energy $\hbar\omega$. Second, superconductivity may provide a matrix element (through particle-hole mixing) that couples a preexisting collective mode to the external probe, magnetic neutron scattering [8].

Further experimental information is clearly necessary in order to distinguish between these fundamentally different interpretations. Since the doping dependence of the superconducting energy gap has recently been determined independently by angle-resolved photoemission [9], the doping dependence of the collective mode may provide additional insights into the mechanism reponsible for coupling of the spin excitations to superconductivity. Further, the normal-state spin susceptibility in $YBa_2Cu_3O_7$ is too small to be reliably determined by present neutron scattering techniques. In underdoped $YBa_2Cu_3O_{6+x}$, however, a sizeable normal-state susceptibility has been observed in previous experiments, so that the relationship between the sharp resonance resonance mode in the superconducting state and the normal-state excitation spectrum can be probed.

The experiments were conducted on the H4M, H7 and H8 thermal triple-axis spectrometers at the High Flux Beam Reactor at Brookhaven and at the SPINS cold-neutron triple axis spectrometer at NIST. At Brookhaven we used neutrons with 30.5 meV final energy and adjusted the energy resolution to $\sim$ 7 meV by collimating the beam. At NIST we used neutrons with 3.5 meV final energy, with typically $\sim$ 0.1 meV energy resolution. In order to convert the observed intensity to the dynamical susceptibility $\chi''(\mathbf{q},\omega)$ in absolute units, the magnetic cross section was calibrated to transverse acoustic phonons around the (004) nuclear Bragg reflection at low energies and to an oxygen vibration of energy 42.5 meV at high energies.

Our primary interest was the influence of superconductivity on the spin excitation spectrum. In Figs. 1 and 2 we have therefore plotted the

*Supported by NSF-DMR94-00362.

Figure 1. Difference of $S(\mathbf{q},\omega)$ (upper panel) and $\chi''(\mathbf{q},\omega)$ (lower panel) at $\mathbf{q} = (\pi,\pi)$ above and below $\mathrm{T_c} = 52\mathrm{K}$ for $\mathrm{YBa_2Cu_3O_{6.5}}$.

Figure 2. Difference of $S(\mathbf{q},\omega)$ (upper panel) and $\chi''(\mathbf{q},\omega)$ (lower panel) at $\mathbf{q} = (\pi,\pi)$ above and below $\mathrm{T_c} = 67\mathrm{K}$ for $\mathrm{YBa_2Cu_3O_{6.7}}$.

difference of the intensity measured at low temperatures below $\mathrm{T_c}$ and the normal-state intensity above $\mathrm{T_c}$ for two $\mathrm{YBa_2Cu_3O_{6+x}}$ crystals of different oxygen concentrations, x $\sim$ 0.5 ($\mathrm{T_c}$=52K) and x $\sim$ 0.7 ($\mathrm{T_c}$=67K). The data were taken at the in-plane wavevector $\mathbf{q} = (\pi,\pi)$, and at the out-of-plane wavevector corresponding to the maximum of the sinusoidal magnetic structure factor. (For further discussions of the structure factor, see, *e.g.*, Ref. [1].) A subset of these data has been reported in Ref. [10], in arbitrary units. The data of Fig. 2 are also consistent with those of Ref. [11]. Here we have plotted the data in two different ways, both as the scattering function $\mathrm{S}(\mathbf{q},\omega)$ which is directly proportional to the magnetic cross section, and as the imaginary part of the dynamical susceptibility which is related to $\mathrm{S}(\mathbf{q},\omega)$ through the fluctuation-dissipation theorem: $\chi''(\mathbf{q},\omega) = [1 - exp(-\hbar\omega/k_B T)]S(\mathbf{q},\omega)$. (The unit conventions are the same as in Ref. [12] and differ by $3\pi\mu_B^2/2$ from those of Ref. [1].)

Clearly, the dynamical susceptibility below $\mathrm{T_c}$ exceeds that above $\mathrm{T_c}$ over a certain energy range, centered around 25 meV for $\mathrm{YBa_2Cu_3O_{6.5}}$ and 33 meV for $\mathrm{YBa_2Cu_3O_{6.7}}$. Fig. 3 shows that for both doping levels the enhancement of the susceptibility is strongly correlated to the onset of superconductivity. For $\mathrm{YBa_2Cu_3O_{6.7}}$ the energy range over which this enhancement is observed is limited only by the experimental resolution. The intrinsic linewidth of the enhanced part of the dynamical susceptibility, $\chi''_+(\mathbf{q},\omega)$, is therefore indistinguishable from zero, as it is for $\mathrm{YBa_2Cu_3O_7}$ where $\chi''_+(\mathbf{q},\omega)$ is centered

Figure 3. Temperature dependence of $\chi''(\mathbf{q},\omega)$ at $\hbar\omega = 25$ meV for $\mathrm{YBa_2Cu_3O_{6.5}}$ (upper panel) and at $\hbar\omega = 33$ meV for $\mathrm{YBa_2Cu_3O_{6.7}}$ (lower panel). The closed (open) symbols represent data taken with a polarized (unpolarized) beam.

Figure 4. $\mathrm{T_c}$-dependence of the enhanced part of the dynamical susceptibility, $\int d\omega \chi''_{+}(\mathbf{q},\omega)$, and the resonance energy $\mathrm{E_{res}}$.

around 40 meV. By contrast, $\chi''_{+}(\mathbf{q},\omega)$ is somewhat broadened in $\mathrm{YBa_2Cu_3O_{6.5}}$.

In Fig. 4 we have summarized our observations. The integrated spectral weight of the enhanced part of the susceptibility, $\int d\omega \chi''_{+}(\mathbf{q},\omega)$, decreases as a function of increasing $\mathrm{T_c}$, whereas $\mathrm{E_{res}}$, the energy around which $\chi''_{+}(\mathbf{q},\omega)$ is peaked, increases. While Fig. 3 shows that the ratio of the resonance spectral weight to the spectral weight of the normal-state spin excitations increases strongly with increasing doping, it is interesting to note that the resonance spectral weight actually decreases on an absolute scale with increasing carrier concentration. The functional dependence of $\mathrm{E_{res}}$ on $\mathrm{T_c}$ (or doping, which depends monotonically on $\mathrm{T_c}$) is obviously not well defined by the three data points, but the qualitative trend contrasts sharply with the weak doping dependence of the energy gap directly determined by photoemission spectroscopy [9]. This discrepancy was predicted in the model of Zhang and collaborators [8] where the resonance energy is not tied to the gap but is directly related to the doping level. It does not rule out the gap model, however. Millis and Monien [5] have shown that the resonance energy can be lower than the gap energy due to final state interactions of the quasiparticle-quasihole pair. These interactions are expected to increase as the doping level is reduced. A more clearcut picture may emerge when the present data are compared in detail to quantitative predictions of the resonance energy and integrated spectral weight [5,8].

Figure 5. Energy integral of the difference of $S(\mathbf{q},\omega)$ above and below T_c at $\mathbf{q} = (\pi,\pi)$ and $0 \leq \hbar\omega \leq 50$ meV, for different doping levels.

Finally, we turn to the relation between the normal-state spin excitations and the resonance in the superconducting state. We are motivated by the total moment sum rule which requires that the the integral $\int d\mathbf{q} d\omega S(\mathbf{q},\omega)$ is weakly temperature dependent (and temperature independent for local-spin models). The difference of $S(\mathbf{q},\omega)$ in the normal and superonducting states is plotted in the upper panels of Figs. 1 and 2 for $\mathbf{q} = (\pi,\pi)$. For $YBa_2Cu_3O_{6.5}$ the positive and negative areas are equal to within our resolution, that is, the total moment sum rule is exhausted for $\mathbf{q} = (\pi,\pi)$ and in the energy range investigated in our experiment ($0 \leq \hbar\omega \leq 50$ meV). By contrast, in both $YBa_2Cu_3O_{6.7}$ and $YBa_2Cu_3O_7$ the positive part of the difference clearly exceeds the negative part in this domain of $(\mathbf{q},\omega)$. This situation is summarized in Fig. 5. For the total moment sum rule to be satisfied, the intensity of the resonance mode must be drawn from a much wider range of $(\mathbf{q},\omega)$. Further investigations, especially at higher energies, are clearly warranted.

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